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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/088,738	07/23/2002	Francis Humblot	33808F172	4589
441	7590	05/03/2006	EXAMINER	
SMITH, GAMBRELL & RUSSELL, LLP			SINGH, PREM C	
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WASHINGTON, DC 20036			PAPER NUMBER	

1764

DATE MAILED: 05/03/2006

Please find below and/or attached an Office communication concerning this application or proceeding.

# Office Action Summary

Application No.

10/088,738

Applicant(s)

HUMBLLOT ET AL.

Examiner

Prem C. Singh

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-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

## Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

## Status

- 1) ☒ Responsive to communication(s) filed on 07 March 2006.
- 2a) ☒ This action is **FINAL**. 2b) ☐ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

## Disposition of Claims

- 4) ☒ Claim(s) 1-24 is/are pending in the application.
- 4a) Of the above claim(s) \_\_\_\_\_ is/are withdrawn from consideration.
- 5) ☐ Claim(s) \_\_\_\_\_ is/are allowed.
- 6) ☒ Claim(s) 1-24 is/are rejected.
- 7) ☐ Claim(s) \_\_\_\_\_ is/are objected to.
- 8) ☐ Claim(s) \_\_\_\_\_ are subject to restriction and/or election requirement.

## Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on \_\_\_\_\_ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.
- Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
- Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

## Priority under 35 U.S.C. § 119

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some \* c) ☐ None of:
1. ☐ Certified copies of the priority documents have been received.
2. ☐ Certified copies of the priority documents have been received in Application No. \_\_\_\_\_.
3. ☒ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

\* See the attached detailed Office action for a list of the certified copies not received.

## Attachment(s)

- 1) ☐ Notice of References Cited (PTO-892)
- 2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
- 3) ☐ Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)  
Paper No(s)/Mail Date \_\_\_\_\_
- 4) ☐ Interview Summary (PTO-413)  
Paper No(s)/Mail Date. \_\_\_\_\_
- 5) ☐ Notice of Informal Patent Application (PTO-152)
- 6) ☐ Other: \_\_\_\_\_

## DETAILED ACTION

### Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

Claims 1-12, and 19-21 are rejected under 35 U.S.C. 103(a) as unpatentable over Zimmermann et al (US Patent 5,922,192) in view of Reed et al (US Patent 5,656,150). The Zimmermann reference discloses a method for treating the heat exchanger surfaces in reactors and/or heat exchangers of installations for converting hydrocarbons and other organic compounds at high temperatures in the gaseous phase (Column 3, lines 20-28). The metallic surfaces coming in to contact with the organic substances are treated at a temperature of 300 to 1000°C, over a period of 0.5 to 12 hours with a mixture of silicon and sulfur containing product and a dry gas flow which is inert with respect to silicon- and sulfur-containing product (Column 3, lines 20-29).

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Zimmermann's prior art describes each and every aspect of the applicant's claim 1 except that the applicant's treatment is done in presence of steam as carrier gas instead of using inert gas as disclosed by Zimmermann. Zimmermann discloses that a carrier gas other than the inert gas for the system can also be used (Column 4, line 64).

Zimmermann explores the possibility of using steam as a carrier gas as disclosed in his example 6 and concludes that steam is not suitable for long lasting suppression of coking on materials pretreated with trimethylsilylmethyl mercaptan (Column 7, lines 45-53). Reed reference teaches a novel method for treating the radiant tubes of a fired pyrolysis heater with an antifoulant composition for inhibiting the formation and deposition of coke thereon (Abstract). Reed's claim #5 states that the diluent fluid is steam (Column 8 lines 51-52).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the process of the Zimmermann reference by using steam as suggested by Reed because of its easy availability especially in the steam cracking units.

Zimmermann reference uses hydrogen, methane, nitrogen, and steam separately as a carrier gas. Although Zimmermann did not use steam and an inert together, it would have been obvious to one having ordinary skill in the art at the time the invention was made to modify Zimmermann's invention to add a step of blending steam and an inert gas as a carrier gas. The purpose of both (steam and inert gas) is as a diluent or a carrier gas (Reed: Column 2, line 30; Zimmermann: column 4, line 64). Since steam and

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inert gas have been proved to be functionally equivalent, blending of the two had a reasonable expectation of success by combining the teachings of Zimmerman and Reed. Zimmermann discloses in his claim #14 wherein the silicon- and sulfur-containing product includes carbon and hydrogen atoms (Column 11, lines 20-22).

Zimmermann teaches the use of several silicon compounds (Column 3, lines 30-35) but does not mention about hexamethyl disiloxane. Reed discloses that out of suitable organic silicon compounds namely: silanes, silicates, and siloxanes, at present hexamethyldisiloxane is preferred (Column 4, lines 61-62).

It would have been obvious to one skilled in the art at the time the invention was made to combine Zimmermann's teachings with Reed's inventions and thereby using hexamethyldisiloxane as a silyl compound.

Zimmermann teaches that silicon- and sulfur-containing product is selected from (1) one or more silicon and sulfur containing volatile compounds, (2) a mixture of silicon containing volatile compounds and a mixture of sulfur containing volatile compounds, and (3) a mixture of silicon-and sulfur-containing volatile compounds (Column 3, lines 30-35). The atomic ratio of silicon to sulfur is 5:1 to 1:1 (Column 3, line 37). Particularly advantageous compounds are trimethylsilyl mercaptan, dimethyl sulfide, dimethyl disulfide, and bis(trimethylsilyl) sulfide and mixtures thereof (Column 3, lines 37-40). Zimmermann discloses that the molar ratio of silicon-and sulfur-containing compounds to the inert gas is between 0.001 and 0.01 (Column 10, lines 60-61). Zimmermann teaches that the pressure of the mixture sent through the system can correspond to the

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usual pressures in the cracking furnace system, e. g. 0.5 to 20 bar, preferably in a range of 1 to 2 bar (Column 4, lines 60-63).

Zimmermann reference teaches that atomic ratio of silicon-and sulfur-containing product is between 5:1 and 1:1 (Column 10, lines 56-57). Zimmermann also discloses that the molar ratio of silicon-and sulfur-containing compounds to the inert gas is between 0.001 and 0.01 (Column 10, lines 60-61). Zimmermann further teaches that the pressure of the mixture sent through the system can correspond to the usual pressures in a cracking furnace system, e.g., 0.5 to 20 bar, preferably in a range of 1 to 2 bar (Column 4, lines 61-63).

Claims 13-18 and 22-24 are rejected as unpatentable over Zimmermann in view of Reed and further in view of Zimmermann et al (US Patent 5,849,176) and Kukes et al (US Patent 4,410,418). Zimmermann's Figure 3 shows the influence of 85 ppm dimethyl disulfide as an addition to n-heptane on the rate of coke formation (Column 5, lines 16-18). Zimmermann has not added a silyl compound in the feedstock of the organic compound to be cracked. Zimmermann (5,849,176) discloses a process for producing thermally cracked products from hydrocarbons while simultaneously, reducing the coke deposits on the heat exchange surfaces by adding to the feed to be cracked, before the cracking temperature is reached, 20 to 1000 ppm of an additive composition that is selected from among one or more volatile organic compounds containing silicon and sulfur (Column 2, lines 27-33).

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It would have been obvious to one skilled in the art at the time the invention was made to combine Zimmermann's teachings with Zimmermann (5,849,176) invention because it has proved advantageous to treat the hydrocarbons to be thermally cracked, before the actual cracking temperature is reached, with an additive that consists either of a volatile compound containing silicon and sulfur in the atomic ratio 5:1 to 1:1 or of a mixture of a compound containing silicon and a compound containing sulfur in the same atomic ratio (Zimmermann- 5,849,176: Column 3, lines 45-51).

Although Zimmermann does not mention about using hexamethyldisiloxane, Zimmermann (5,849,176) discloses that the compound containing silicon and/or sulfur is preferably selected from the group that consists of trimethyl silyl mercaptan, dimethyl sulfide, tetra methyl silane and bis trimethyl silyl sulfide and their mixtures. However, other volatile compounds can also be used (Column 2, lines 65-67; Column 3, lines 1-2). Kukes discloses a method for reducing the formation of coke on the metals which are contacted with a gaseous stream containing hydrocarbons in a thermal cracking process by contacting said metals with a halogen containing silicon compound from the group consisting of halogen containing silanes, disilanes, and siloxanes (Column 8, lines 5-13).

It would have been obvious to one skilled in the art at the time the invention was made to combine Zimmermann's teachings with Zimmermann (5,849,176) and Kukes inventions because the use of hexamethyl disiloxane is preferred over other silicon compounds.

Zimmermann's Figure 3 shows the influence of adding dimethyl disulfide to the cracking stock (n-heptane) on the rate of coke formation. Although Zimmerman does not show the effect of adding silicon to the cracking stock, Zimmermann (5,849,176) discloses to treat the hydrocarbons to be cracked before the actual cracking temperature is reached, with an additive that consists silicon and sulfur in the atomic ratio 5:1 to 1:1 (Column 3, lines 45-50). In his laboratory experiment of Example 10, PEP 5, Zimmermann (5,849,176) mentions that 100 ppm of an equimolar mixture of tetra-methyl-silane and dimethyl sulfide in n-heptane are taken for cracking reactions (Column 7, lines 27-28).

It would have been obvious to one skilled in the art at the time the invention was made to have modified the process of Zimmermann reference by including silicon with sulfur as suggested by Zimmermann (5,849,176) because the use of silicon and sulfur is preferred for reduction of coking.

Although Zimmermann uses only sulfur (dimethyl disulfide) in the n-heptane cracking stock (Column 5, lines 16-17), Zimmermann (5,849,176) teaches the use of silicon and sulfur in the atomic ratio 5:1 to 1:1 (Column 3, lines 45-50) and mentions that 100 ppm of an equimolar mixture of tetra-methyl-silane and dimethyl sulfide in n-heptane are taken for cracking reactions (Column 7, lines 27-28).

It would have been obvious to one skilled in the art at the time the invention was made to combine Zimmermann's invention with Zimmermann (5,849,176) and Kukes to obtain the desired concentration and ratio of silicon and sulfur.



***Response to Arguments***

The amendments to claims 1, 6- 9 are noted.

The applicant has added the limitation of using non-silicon sulfur and non-sulfur silicon.

Zimmermann invention discloses the use of compounds including dimethyl sulfide and dimethyl disulfide (Column 3, lines 37-39). These two are non-silicon sulfur compounds.

Reed invention discloses the use of various compounds (Column 4, lines 50-60). They are non-sulfur silicon compounds.

The applicant's argument states that Zimmermann does not teach using silicon or sulfur compounds with steam and Reed invention does not teach or suggest the use of steam with an antifoulant composition containing a silicon compound and a sulfur compound.

Zimmermann's Example 2 uses dimethyl disulfide, a compound which is known and used industrially as a coking inhibitor (Column 6, lines 44-46). Table 1 shows results of using (d) 2:1, (e) 3:1, (f) 4:1, and (g) 5:1 mixtures of tetramethyl silane and dimethyl sulfide (Column 10, lines 14-17). Zimmermann uses nitrogen as carrier but suggests that a carrier gas other than the inert gas for the system can also be used (Column 4, lines 64-65).

Reed invention discloses using silicon compounds (Column 4, lines 50-60), and tin compounds having sulfur (Column 5, lines 11-13; lines 26-30) with steam (Column 6,

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lines 8-22). Reed presents an example where the treating agent is tetrabutyl tin (no silicon or sulfur).

Kukes invention discloses that a preferred pretreatment method is to contact the metals with a silicon compound by flooding the cracking tubes with silicon compound (Column 3, lines 59-61). Any suitable solvent may be used to form a solution of the silicon compound. The examples of suitable solvents are carbon tetrachloride, benzene, and carbon disulfide (Column 4, lines 15-19). Steam is generally utilized as a diluent for the hydrocarbon containing feedstock flowing to the cracking furnace (Column 4, lines 64-66).

Thus, the application of non-sulfur silicon compounds and non-silicon sulfur compounds using steam to inhibit coking is considered prima facie obvious in view of combined teachings of Zimmermann, Reed, and Kukes. Nothing unexpected is evidenced to overcome this prima facie case.

### **Conclusion**

**THIS ACTION IS MADE FINAL.** Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire **THREE MONTHS** from the mailing date of this action. In the event a first reply is filed within **TWO MONTHS** of the mailing date of this final action and the advisory action is not mailed until after the end of the **THREE-MONTH** shortened statutory period, then the

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shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of this final action.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Prem C. Singh whose telephone number is 571-272-6381. The examiner can normally be reached on MF 6:30-3:00.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Glenn Caldarola can be reached on 571-272-1444. The fax phone number for the organization where this application or proceeding is assigned is 703-872-9306.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

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